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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁶ : B01J 19/10	A1	(11) International Publication Number: WO 96/20784 (43) International Publication Date: 11 July 1996 (11.07.96)
<p>(21) International Application Number: PCT/US96/00466</p> <p>(22) International Filing Date: 4 January 1996 (04.01.96)</p> <p>(30) Priority Data: 08/368,816 6 January 1995 (06.01.95) US</p> <p>(71) Applicant: TRUSTEES OF BOSTON UNIVERSITY [US/US]; 147 Bay State Road, Boston, MA 02215 (US).</p> <p>(72) Inventor: MADANSHETTY, Sameer, I.; 36 Armory Street, Cambridge, MA 02139 (US).</p> <p>(74) Agents: KLOCINSKI, Steven, P. et al.; Baker & Botts, L.L.P., The Warner, 1299 Pennsylvania Avenue, N.W., Washington, DC 20004 (US).</p>		<p>(81) Designated States: AL, AM, AT, AU, AZ, BB, BG, BR, BY, CA, CH, CN, CZ, DE, DK, EE, ES, FI, GB, GE, HU, IS, JP, KE, KG, KP, KR, KZ, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TR, TT, UA, UG, UZ, VN, ARIPO patent (KE, LS, MW, SD, SZ, UG), Eurasian patent (AZ, BY, KZ, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).</p> <p>Published <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i></p>
<p>(54) Title: METHOD AND APPARATUS FOR ENHANCING CHEMICAL REACTIONS THROUGH ACOUSTIC CAVITATION</p> <p>(57) Abstract</p> <p>The invention relates to a method and apparatus for catalyzing reactions through the inducement of acoustic cavitation. Cavitation is induced by applying ultrasonic energy of two separate frequencies to a liquid reaction medium. A cavitation field having relatively high tensile pressure and a coaxing field of higher frequency and lower tensile pressure than the cavitation field are applied to a liquid reaction medium to produce an amount of cavitation suitable for catalyzing chemical reactions.</p>		

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METHOD AND APPARATUS FOR ENHANCING CHEMICAL REACTIONS THROUGH ACOUSTIC CAVITATION

BACKGROUND

5 Field of the Invention

This invention relates to the field of sonochemistry. More specifically, this invention is directed to the catalysis of chemical reactions through the use of enhanced acoustic microcavitation.

Background of the Invention

10 Cavitation is the formation and subsequent implosion of cavities (bubbles) in a liquid when subjected to a certain level of vibration. Cavitation generally results from a pre-existing gaseous presence, either as a bubble or a stabilized pocket of gas trapped in a crevice (such a crevice could be found in a container wall or on a fluid-borne particle). When exposed to vibration, the
15 gas may release and the resulting bubble may implode. An ultrasonic wave has alternating positive (high) and negative (low) half-cycles which may cause the necessary vibration and induce cavitation.

The mechanism responsible for acoustic cavitation can be understood by considering a free bubble in the path of a sound wave. The
20 bubble expands and contracts in response to the alternating negative and positive half-cycles of the sound wave respectively. Energy is stored during the expansion and released in concentrated form during the possibly implosive collapse. Should a bubble grow to about two and a half times its original size during the negative half-cycle of the wave, then during the following positive
25 half-cycle its speed of collapse could become supersonic. The occurrence of these single cycle violent events is referred to as transient cavitation.

There also exists a more gradual process, termed rectified diffusion which may lead to cavitation. Under favorable conditions, a small bubble exposed to a continuous sound wave tends to grow in size if rectified
30 diffusion is dominant. According to Henry's law, for a gas soluble in liquid, the

equilibrium concentration of the dissolved gas in the liquid is directly proportional to the partial pressure of the gas above the liquid surface, the constant of proportionality being a function of temperature only. When a bubble expands, the pressure in the bubble interior falls and gas diffuses into the bubble from the surrounding liquid. When the bubble contracts the pressure in the interior increases and the gas diffuses into solution in the surrounding liquid. However, the area available for diffusion is larger in the expansion mode than in the contraction mode. Consequently, there is a net diffusion of the gas into the bubble from the surrounding liquid over a complete cycle and thus the bubble grows due to rectified diffusion.

However, a bubble undergoing rectified diffusion in the path of a sound wave can grow only up to a critical size, its resonance radius. The resonance radius is a function of the frequency of the sound wave. There is a simple relation for the resonance radius of air bubbles in water:

$$\text{(resonance radius in } \mu\text{m)} \times \text{(insonification frequency in MHz)} = 3.2$$

This relation is valid to within 5% even for bubbles with radii as large as 10 μm . When a bubble reaches its resonance radius through rectified diffusion, its response to the sound wave becomes increasingly vigorous. A post-resonance bubble may either exhibit nonlinear modes of oscillations or become transient if the forcing acoustic pressure amplitude is adequately high.

It is important to note that at high frequencies it becomes increasingly difficult to bring about cavitation because insufficient time is available for bubble growth, which precedes the cavitation implosion. Consequently, the cavitation threshold of bubbles becomes greater at higher frequencies, and for practical purposes it becomes very difficult to bring about cavitation (in water) at frequencies in excess of 5 MHz.

Sonochemistry generally refers to the use of acoustic energy to enhance and accelerate (i.e. catalyze) chemical reactions especially organic syntheses. In particular, an ultrasonic field may be used to catalyze a reaction by inducing microcavitation. The catalysis effect of cavitation is thought to be
5 do to, *inter alia*: localized stirring; the creation of "hot spots" (do to the fact that the collapse of a bubble occurs adiabatically); sharp shear gradients; and, high pressure points.

Generally, a certain threshold level of cavitation activity is required in order to give rise to the catalysis effect. Heretofore, efforts to
10 induce cavitation in a liquid have been concentrated around the use of a single ultrasound source having a frequency of about 20 kHz. This source, generally known as a sonicator, was generally employed in the laboratory environment, and produced cavitation which was erratic and could not be controlled. These previous efforts to use acoustically induced cavitation to catalyze reactions have
15 been ineffective especially in regard to scale-up, i.e. converting laboratory experiments to industrial applications. They have failed to produce the necessary level of cavitation efficiently enough so as to warrant its use as a catalyst. Further, when cavitation was produced, it was largely unpredictable and therefore could not be consistently reproduced or controlled.

20

SUMMARY OF THE INVENTION

Therefore, a need has arisen for a method and apparatus which can efficiently produce an amount of cavitation necessary to effectively and controllably catalyze chemical reactions in a liquid medium.

Accordingly, it is an object of the present invention to provide a
25 method and an apparatus for efficiently producing an amount of cavitation necessary to catalyze chemical reactions in a liquid medium.

It is a further object of the present invention to provide a method and an apparatus which can consistently produce and control an amount of

cavitation necessary to catalyze chemical reactions in a liquid medium using acoustic energy.

The objects of the present invention are generally achieved by inducing cavitation through the use of two overlapping ultrasonic fields.

5 In one embodiment, gas caps are formed on particles present in a liquid medium by application of an ultrasonic field having relatively high frequency and relatively low pressure. The gas caps are formed in part by the agglomeration of nano-gas-dots, which are extremely small gas pockets present on the particles. Cavitation is then induced by separating the gas caps from the
10 particles by application of a second ultrasonic field having relatively low frequency and relatively high pressure.

Further, an apparatus according to the present invention includes a first ultrasound source generating a relatively high frequency field and a second ultrasound source generating a relatively low frequency field arranged
15 so that the resulting high and low frequency fields overlap when applied to the liquid medium.

In a modified embodiment, the gas cap formation may be accomplished through the application of plural relatively high frequency ultrasonic fields. Cavitation is then induced by application of at least one
20 relatively low frequency ultrasonic field to separate the gas caps from the particles. Each high frequency field overlaps a relatively low frequency ultrasonic field.

An apparatus according to the modified embodiment could include a plurality of relatively high frequency ultrasound sources and at least one
25 relatively low frequency ultrasound source. The sources could be arranged so that the fields resulting from the relatively high frequency sources and the field resulting from the at least one relatively low frequency source overlap when applied to the liquid.

In a further modification, the relatively high frequency fields and the relatively low frequency fields in any of the above embodiments are arranged so that they are confocal.

BRIEF DESCRIPTION OF THE DRAWINGS

5 Figure 1 depicts a flow diagram of one embodiment of a method for enhancing chemical reactions through acoustic cavitation.

 Figure 2(a) depicts a schematic representation of a particle in a liquid having nano-gas-dots disposed thereon.

 Figure 2(b) depicts a schematic representation of a particle in a
10 liquid during formation of gas caps on the particle.

 Figure 2(c) depicts a schematic representation of a particle and two bubbles in a liquid produced through cavitation.

 Figure 3 depicts a schematic representation of one embodiment of an apparatus for catalyzing a chemical reaction using acoustic cavitation.

15 Figure 4 depicts a schematic representation of another embodiment of an apparatus for catalyzing a chemical reaction using acoustic cavitation.

 Figure 5(a) depicts a schematic representation of a side view of another embodiment of an apparatus for catalyzing a chemical reaction through
20 acoustic cavitation.

 Figure 5(b) depicts a schematic representation of a cross section of the embodiment of an apparatus for catalyzing a chemical reaction shown in figure 5(a).

DETAILED DESCRIPTION

25 The present invention utilizes cavitation to catalyze reactions in a liquid medium. Clean liquids do not cavitate easily. A pure liquid purged of all particulate impurities and stored in a perfectly smooth container can attain its theoretical tensile strength before undergoing cavitation or fracture. Under ideal

conditions, the theoretical tensile strength of water based on the homogeneous nucleation theory exceeds 1000 bars (1 bar is approximately equal to atmospheric pressure at 15 degrees centigrade and 1 atmosphere, which is approximately equal to 10^5 Newtons per square meter). In cavitation studies

5 tensile strengths of liquids are often quoted in terms of negative pressures (bar peak negative). A liquid's "threshold" is understood as the pressure amplitude at which the first occurrence of cavitation is detected.

In practice, observed thresholds of clean liquids are generally lower than those theoretically possible. Rarely does the strength of even a

10 reasonably clean liquid exceed a few bars, primarily because gas pockets exist within liquids which provide the necessary seeding for cavitation. A gas pocket is often stabilized in a crevice, either in the container wall or on a liquid-borne particle. Incomplete wetting traps gas at the root of a sharp crevice, stabilizing it against dissolution. Unlike a free bubble, surface tension in this case acts on

15 a meniscus which is concave towards the liquid. Over-pressuring the liquid for sufficient duration prior to insonification can force the meniscus further into the crevice and may cause full wetting of the crevice, which then gives rise to increased thresholds.

The present invention allows the threshold at which cavitation is

20 induced to be lowered by application of a high frequency field. The lowering of the threshold allows cavitation to be produced in a more efficient manner. This in turn allows the amount of cavitation to be controlled so that cavitation can be effectively used to catalyze reactions in a liquid medium.

Fig. 1, depicts a flow diagram of one embodiment of a method

25 for enhancing chemical reactions through acoustic cavitation. The method for enhancing chemical reactions according to this embodiment catalyzes the reaction between a combination of reactants in a liquid medium. In initial step 100, reactants are combined in a liquid medium. A high frequency acoustic field

which induces gas caps to form on particles in a liquid is generated in a step 200. Next, in step 300, particles having gas caps formed thereon are subjected to a cavitation field to separate the gas caps from the particles thereby inducing cavitation. In step 400, the induced cavitation catalyzes the reaction through
5 localized stirring, the creation of "hot spots" (do to the fact that the collapse of a bubble occurs adiabatically), the creation of sharp shear gradients and the creation of high pressure points. Steps 200-400 will each be explained in greater detail below.

In step 200, a high frequency acoustic field is used to induce the
10 formation of gas caps. The high frequency field will preprocess the liquid medium for cavitation by inducing gas caps to form on particles in the liquid. That is, the high frequency field "coaxes" cavitation in the liquid and therefore can be thought of as a coaxing field. The high frequency field generally has a frequency greater than 5 MHz. The high frequency field is applied with a
15 relatively low pressure on the order of 0.5 bar peak negative. A 30 MHz, 0.5 bar peak negative focused acoustic field may be used as the high frequency field.

Application of the high frequency field to the reaction liquid will cause gas pockets to form on particles in the liquid. These pockets of gas are referred to as gas caps. Gas cap formation is made possible by the existence of
20 very small gas dots on the surface of the particles. Solids, such as particles, disposed in liquid have at least microscopic surface areas which do not become wetted. Very few solids have geometrically smooth surfaces. Solids in glass state may have smooth surfaces, and carefully cleaved crystal surfaces may be smooth initially. But, the notion of smoothness for real surfaces implies that
25 roughness below a certain length scale can be ignored. If even fine length scale matters are considered, then roughness persists at some level. Adsorption studies on various powder surfaces have revealed the presence of fractal surfaces even to the molecular levels. At some scale of fineness there must be

unwet sites on the solid surface. Wetting does not occur on the entire solid surface because no solid has a geometrically smooth surface when viewed at the microscopic level. Because liquids operate to minimize surface energy which minimizes free surfaces, these microscopic areas of roughness trap gas therein, and thus, do not become wetted with liquid. These areas of roughness, however, are microscopic and believed to be at the nanometer length scale. Gas trapped in the areas of roughness are thus referred to as nano-gas-dots. Nano-gas-dots may be formed by gas released in the dissolved state, vapor molecules from the host liquid, and under favorable circumstances, desorption of adsorbed non-polar constituents. These nano-gas-dots will coalesce and grow under the influence of the high frequency field and thereby form the gas caps. These processes of coalescence and growth will now be explained.

Referring to Fig. 2(a), particle 50 in liquid 70 is depicted as having a plurality of nano-gas-dots 55 disposed thereon. As discussed above, the size of nano-gas-dots 55 is very small and thus the depiction in Fig. 2(a) is disproportionate only for purposes of illustration.

When a sound wave, such as that generated by the high frequency acoustic field, is introduced into the liquid, all of the contents of the liquid oscillate in response. A particle in the liquid which is heavier than the liquid oscillates with an amplitude less than that of the liquid. A particle which is lighter than its surrounding liquid oscillates with a greater amplitude than that of the liquid.

In general, the density of particle 50 does not differ significantly from that of liquid 70, and consequently particle 50 follows the oscillation path of liquid 70. However, the density of air is generally much less than that of the liquid (a factor of 830 less if the liquid is water). Therefore, nano-gas-dots 55 on the surface of particle 50, have an amplitude of oscillation greater than that of liquid 70 (approximately three times greater if the liquid is water). This

difference in amplitude of oscillation is significant because it is one of the factors which leads to gas cap formation. That is, the oscillations may cause nano-gas-dots to bridge with neighboring dots due to the attraction forces between them. The attraction forces are due to hydrodynamic interaction between the particles.

- 5 For example, two pulsating hemispherical nano-gas-dots closely separated by 50 nanometers on a plane such as a particle surface having a one micron diameter spherical shape, may coalesce as a result of mutual attraction.

Another factor which may lead nano-gas-dots to coalesce and form gas caps is that the density of the air will generally be much less than the
10 density of the host liquid. Because of this density contrast, the nano-gas-dots will experience a greater amount of kinetic buoyancy than the particles. Therefore, the movement of the nano-gas-dots will generally lead that of the particle in response to an incident wave. This lead will urge the nano-gas-dots toward the fore and aft regions of the particle and will therefore increase the
15 chances of the nano-gas-dots coalescing at the fore and aft regions.

An illustration of how this density difference effects gas cap coalescence can be explained with reference to Fig. 2(b). Consider a nano-gas-dot midway between X and Y at approximately 30° latitude (where the Y-Y line is the equator). The high frequency field will cause this gas dot to accelerate
20 alternately in the directions indicated by arrows A and B. During the phase of acceleration indicated by arrow A, this nano-gas-dot will be free to move towards Y. During the opposite half cycle of the high pressure wave, this acceleration will be reversed to the direction of arrow B. When this occurs, the nano-gas-dot will encounter a material barrier due to the particle that resists the
25 body force of the kinetic buoyancy. Under these circumstances, the nano-gas-dot may be thought to be experiencing a rectified kinetic buoyancy, which promotes the agglomeration of nano-gas-dots at fore and aft regions along the

axis of oscillation and the consequent formation of gas caps. That is, the nano-gas-dot will experience a ratcheted motion toward Y.

The above described mechanisms explain how the high frequency field can lead to the coalescence of nano-gas-dots 55 on the surface of particle 50 at opposite poles along the axis of oscillation on particle 50 as depicted in Fig. 2(b). However, gas cap growth need not occur solely through coalescence.

Gas caps 60 and 80 may also be enlarged due to an infusion of gas from the surrounding liquid. After the phase of acceleration in the direction indicated by arrow B, the liquid in the vicinity of the particle around Y will appear supersaturated with dissolved gas. Dissolved gas generally moves toward the direction of acceleration (as is illustrated in the case of a centrifuge where dissolved gases move toward the center of rotation). This will cause the dissolved gas to supersaturate the liquid next to the gas cap at Y, facilitating gas transfer into the gas cap. This transfer of gas into the gas cap will lead to further enlargement of the gas caps 60 and 80.

Gas caps 60 and 80 tend to be crescent shaped due to the tensile field around the particle. Gas caps 60 and 80 may also be partially vaporous even though nano-gas-dots 55 are unlikely to contain significant amounts of vapor. Gas caps 60 and 80 maintain contact with particle 50 due to surface tension. This surface tension is generally strong enough to maintain contact with gas caps 60 and 80 up to a critical gas cap size. To separate a gas cap from the particle, the tensile pressure on the gas cap due to the acoustic wave must overcome the surface tension resistance along the perimeter of contact. The peak negative pressure amplitude which causes this separation is the cavitation threshold. By causing the gas caps to form and grow on the surface of a particle, the cavitation threshold is thus lowered. When separation occurs, gas caps 60 and 80 break away from particle 50 to form bubbles 90 and 95 as shown in Figure 2(c).

The application of the high frequency field will not alone cause cavitation to occur. One reason is that, the high frequency field does not provide sufficient time for bubble growth. Additionally, the high frequency field is not of sufficient tensile pressure to overcome the surface tension effects and separate a gas cap from a particle or to cause collapse of the bubble. Note that these factors all relate to the size of the bubble. A brief analysis may help explain this. Although gas caps 60 and 80 are not perfect bubbles, because only minor differences in analysis exist between partial bubbles and bubbles, gas caps 60 and 80 may be analyzed as bubbles.

Every bubble has a radius at which forces tending to expand the bubble overcome forces trying to collapse the bubble. At that radius, known as the critical radius, the bubble experiences unbounded increase in size until it fractures. The Blake threshold is the acoustic pressure amplitude that causes the bubble radius to expand beyond the critical radius. The equation for the Blake threshold is given below:

$$R_c = \frac{4}{3} \frac{\sigma}{p_v - p_{lc}} \quad (1)$$

where R_c is the critical radius, σ is the surface tension of the water, p_v is the pressure of the vapor in the bubble, and p_{lc} is the acoustic pressure of the surrounding liquid. Assume that for the gas caps forming on particles in a liquid, the size of the gas cap can be approximated by the size of the particle. In this case, for particles having diameters of $0.984\mu\text{m}$, $0.481\mu\text{m}$, and $0.245\mu\text{m}$, the Blake thresholds are 1.96, 3.1 and 5.3 bar peak negative, respectively. Therefore, not only does a high frequency acoustic field not afford enough time for significant bubble growth, but the maximum pressure amplitude available in

the high frequency acoustic field used in step 200 of about 0.5 bar peak negative is well below the Blake threshold value necessary to cause unbounded growth in the gas caps. Note also that the assumption that the gas cap size is the particle size is actually a conservative assumption in this case because the gas cap size is unlikely to be greater than the particle size and a smaller gas cap size would yield a greater Blake threshold.

It can be seen from this analysis that the high frequency acoustic field pre-processes the particles for cavitation by engendering the formation of gas caps and thereby lowering the cavitation threshold. The high frequency field is not able to wrench off the gas caps because the gas caps do not grow to a size large enough for the relatively low pressure available in the high frequency field to overcome the surface tension on the particle. Therefore, in step 300, a cavitation field is used to focus a stronger tensile pressure toward the particles to cause the gas caps to reach the critical radius and thus separate from the particle.

In step 300 of Figure 1, the particles are subjected to a cavitation field in order to separate gas caps 60 and 80 from particle 50 to form respective bubbles 90 and 95, shown in Figure 2(c). The cavitation field is a relatively low frequency field (compared to the high frequency field). The cavitation field may have a frequency less than 5 MHz and generally within the range 0.5 MHz. - 3 MHz. The cavitation field is applied with a tensile pressure appropriate for overcoming the cavitation threshold. This tensile pressure could be as low as 5 bar peak negative and is preferably within the range of 5 - 7 bars peak negative.

The steps 200 and 300 although shown in series in Figure 1, are generally accomplished simultaneously. That is, a high frequency field and a cavitation field are applied simultaneously to the liquid reaction medium. The two fields can be confocal or they may overlap.

In the final step 400, the separation of the bubbles gives rise to cavitation. This cavitation in the reaction chamber gives rise to a catalysis effect. The cavitation may catalyze reactions in many ways. For instance, during cavitation, a bubble may collapse adiabatically giving rise to a "hot spot."

- 5 These "hot spots" may give rise to a catalysis effect in certain reactions. The cavitation may cause local stirring which also will give rise to a catalysis effect in certain reactions. The cavitation may cause sharp shear gradients at a bubble wall during implosion. These sharp shear gradients may also catalyze certain reactions. For example, the shear gradient may be great enough to catalyze a
- 10 depolymerization by providing sufficient force to snap a long chained polymer in the vicinity of the bubble wall. Cavitation may also create high pressure points during bubble implosion which can also catalyze some reactions.

- The method outlined above also allows control of the cavitation produced and therefore control of the catalysis effect. As noted above it is the
- 15 overlapping of the high frequency field and the cavitation field which causes the cavitation to occur. More specifically, application of the high frequency field lowers the cavitation threshold to the point where cavitation can be caused by a cavitation field having a lower frequency than would normally be required. Because two sources are used it becomes a simple matter to control the amount
- 20 of cavitation produced by controlling the high frequency source. For instance, if both sources are operating continuously, the cavitation produced may become too vigorous. If this occurs, the high frequency source could simply be intermittently turned off which would lead to a rise in the cavitation threshold and a commensurate reduction in the amount of cavitation produced. This
- 25 control could also be accomplished by operating the high frequency source in a pulse mode. If the cavitation became too vigorous, the duty cycle of the high frequency source could be lowered thereby leading to an effective increase in the

cavitation threshold. If cavitation was too sluggish, the duty cycle could be increased.

Having described an embodiment of the method of the present invention, an embodiment of an apparatus for carrying out the present invention
5 will now be explained in conjunction with Figure 3.

The apparatus for carrying out the present invention shown in Figure 3 includes a reaction chamber 30 suitable for carrying out a reaction to be catalyzed. The reaction chamber may be any defined space including but not limited to a tank, vessel or pipe wherein the reactants will be either partially or
10 fully combined. The reaction chamber is filled with a liquid within which the reaction will take place. The cavitation is generally produced in a zone 40 where a high frequency field 50 overlaps with a cavitation field 60.

The high frequency field 50 is generated by a high frequency source 10 which may be an active transducer. The high frequency field is
15 generally of a frequency above 5 MHz and generally has a pressure of about 0.5 bar peak negative. In one embodiment, the high frequency field may be a 30 MHz, 0.5 bar peak negative field. The source 10 may be a 30 MHz transducer providing a focused field. The source 10 may operate in either a continuous or pulse mode and could be a PZT, PZT-5, PVDF or any piezo-ceramic active
20 transducer. Transducers employing lithium-niobate can also advantageously be employed. Active transducers made by Harrisonics, EBL, or Panametrics could be used as the source 10. Other high frequency, low pressure fields may also be used.

In Figure 3, the cavitation field and the high frequency field are
25 shown in relatively confocal arrangement. This does not have to be the case because the two fields need only overlap. The cavitation field 60 is generated by cavitation source 20. The cavitation field is generally a low frequency high pressure field as compared to the high frequency field. It generally has a

frequency below 5 MHz and preferably in the range 0.5 MHz - 3 MHz. As explained above, the cavitation field is applied with a pressure suitable to overcome the reduced cavitation threshold and may be as low as 5 bar peak negative. In one embodiment, the cavitation field may have a frequency of .75
5 MHz and a pressure of 7 bar peak negative. The cavitation source 20 may be PZT-5 transducer. The cavitation source 20 could also be an LZT-1 transducer. The cavitation source can operate in either a continuous or pulse mode and preferably will provide a focused or slightly focused field (although an unfocused field could be used for some specific applications).

10 In the embodiment shown in Figure 3, the overlapping of the high frequency field and the cavitation field will lead to the formation of gas caps and the inducing of cavitation as explained with regard to Figures 1 and 2.

As explained above, the overlapping of a high frequency and a cavitation field will lead to increased cavitation and catalysis of chemical
15 reactions occurring in liquid media. The present invention is not limited however to the use of a single high frequency source and a single cavitation source. Multiple high frequency sources and cavitation sources could be overlapped to produce an even greater amount of cavitation with a commensurate catalysis effect. One embodiment including an alternative arrangement of
20 sources is shown in Figure 4.

Figure 4 shows a reaction chamber 30 suitable for carrying out a reaction to be catalyzed according to the present invention. The reaction chamber 30 has high frequency sources 10 and cavitation sources 20 arranged on each side. Although Figure 4 does not show the specific fields, it is apparent
25 that such an arrangement of sources would provide four zones where the fields overlap thereby engendering gas cap formation and cavitation according to the method described in conjunction with Figures 1 and 2.

Figures 5 show another embodiment of an apparatus for catalyzing chemical reactions using acoustic cavitation. In this embodiment the reaction chamber is a pipe 35 through which there will be a flow of reactants in a liquid medium. Figure 5(a) shows a side view of a pipe 35 containing a flow
5 F of reactants in a liquid medium. A set of sources, i.e. a high frequency source 10 and a cavitation source 20, are arranged around the periphery of the pipe separated by an angle A of approximately 90 degrees as shown in Figure 5(b). A plurality of these sets of sources can be arranged along the length of the pipe as shown in figure 5(a). The frequencies and pressures of the various sets of
10 sources as well as their operational mode (i.e. continuous or pulse) could then be tuned to provide for different levels of cavitation at different points along the length of the pipe.

Figures 4 and 5 are merely examples of possible arrangements of sources for inducing cavitation. Any number of sources could be used in any
15 arrangement as long as there is some overlap of a high frequency field and a cavitation field. Further, there does not need to be a one to one correspondence between the number of high frequency sources and cavitation sources. It is possible to use a number of high frequency sources with a single cavitation source to provide a field which overlaps the fields produced by the high
20 frequency sources.

Although a detailed description of the present invention has been provided, it should be understood that the scope of the invention is not to be limited thereby, but is to be determined by the claims which follow. Various modifications and alternatives will be readily apparent to one of ordinary skill
25 in the art.

CLAIMS

We claim:

1. A method for catalyzing reactions in a liquid medium including particles comprising the steps of:
 - 5 combining reactants necessary for performing a chemical reaction in the liquid medium; and,
 - inducing cavitation in said liquid medium using ultrasonic energy of two frequencies;
 - wherein said induced cavitation catalyzes said chemical reaction.
- 10 2. The method of claim 1 wherein the step of inducing cavitation comprises the steps of:
 - forming gas caps on said particles; and,
 - separating said gas caps from said particles.
3. The method of claim 2 wherein the step of forming gas caps
15 comprises the steps of:
 - generating at least one high frequency ultrasonic field having a frequency higher than about 5 MHz and a pressure of about 0.5 bar peak negative; and,
 - applying said at least one high frequency ultrasonic field to the
20 liquid medium.
4. The method of claim 3 wherein the step of separating said gas caps from said particles comprises the steps of:
 - generating at least one low frequency ultrasonic field having a frequency substantially within the range of about 0.5 MHz - 3 MHz and a
25 pressure generally higher than about 5 bar peak negative; and,
 - applying said at least one low frequency ultrasonic field to said liquid medium.

5. The method of claim 4 wherein the step of applying at least one low frequency ultrasonic field and the step of applying at least one high frequency ultrasonic field are accomplished so that one low frequency ultrasonic field and one high frequency ultrasonic field are applied in an overlapping manner.

6. The method of claim 4 wherein the step of applying at least one low frequency ultrasonic field and the step of applying at least one high frequency ultrasonic field are accomplished so that one low frequency ultrasonic field and one high frequency ultrasonic field are applied confocally.

7. The method of claim 1 wherein the step of inducing cavitation in said liquid medium comprises the steps of:

generating at least one high frequency ultrasonic field having a frequency greater than about 5 MHz and a pressure of about 0.5 bar peak negative;

generating at least one low frequency ultrasonic field having a frequency substantially within the range of about 0.5 MHz - 3 MHz and a pressure generally higher than about 5 bar peak negative; and,

applying one of said high frequency and one of said low frequency ultrasonic fields to said liquid medium.

8. The method of claim 7 wherein the step of applying comprises the step of applying one of said high frequency and one of said low frequency ultrasonic fields in an overlapping manner.

9. The method of claim 7 wherein the step of applying comprises the step of applying one of said high frequency and one of said low frequency ultrasonic fields confocally.

10. An apparatus for catalyzing reactions in a liquid medium including particles comprising:

a chamber for combining reactants necessary for performing a chemical reaction in the liquid medium; and,

means for inducing cavitation in said liquid medium using ultrasonic energy of two frequencies;

5 wherein said means for inducing cavitation catalyzes said chemical reaction.

11. The apparatus of claim 10 wherein the means for inducing cavitation comprises:

 means for forming gas caps on said particles; and,

10 means for separating said gas caps from said particles.

12. The apparatus of claim 11 wherein the means for forming gas caps comprises:

 means for generating at least one high frequency ultrasonic field having a frequency higher than about 5 MHz and a pressure of about 0.5 bar
15 peak negative; and,

 means for applying said at least one high frequency ultrasonic field to the liquid medium.

13. The apparatus of claim 12 wherein the means for separating said gas caps from said particles comprises:

20 means for generating at least one low frequency ultrasonic field having a frequency substantially within the range of about 0.5 MHz - 3 MHz and a pressure generally higher than about 5 bar peak negative; and,

 means for applying said at least one low frequency ultrasonic field to said liquid medium.

25 14. The apparatus of claim 13 wherein the means for applying at least one low frequency ultrasonic field and the means for applying at least one high frequency ultrasonic field operate so that one low frequency ultrasonic field and one high frequency ultrasonic field are applied in an overlapping manner.

15. The apparatus of claim 13 wherein the means for applying at least one low frequency ultrasonic field and the means for applying at least one high frequency ultrasonic field operate so that one low frequency ultrasonic field and one high frequency ultrasonic field are applied confocally.

5 16. The apparatus of claim 10 wherein the means for inducing cavitation in said liquid medium comprises:

means for generating at least one high frequency ultrasonic field having a frequency greater than about 5 MHz and a pressure of about 0.5 bar peak negative;

10 means for generating at least one low frequency ultrasonic field having a frequency substantially within the range of about 0.5 MHz - 3 MHz and a pressure generally higher than about 5 bar peak negative; and,

means for applying one of said high frequency and one of said low frequency ultrasonic fields to said liquid medium.

15 17. The apparatus of claim 16 wherein the means for applying comprises means to apply one of said high frequency and one of said low frequency ultrasonic fields in an overlapping manner.

18. The apparatus of claim 16 wherein the means for applying comprises means to apply one of said high frequency and one of said low
20 frequency ultrasonic fields confocally.

19. The apparatus of claim 10 wherein the chamber for combining reactants comprises a pipe.

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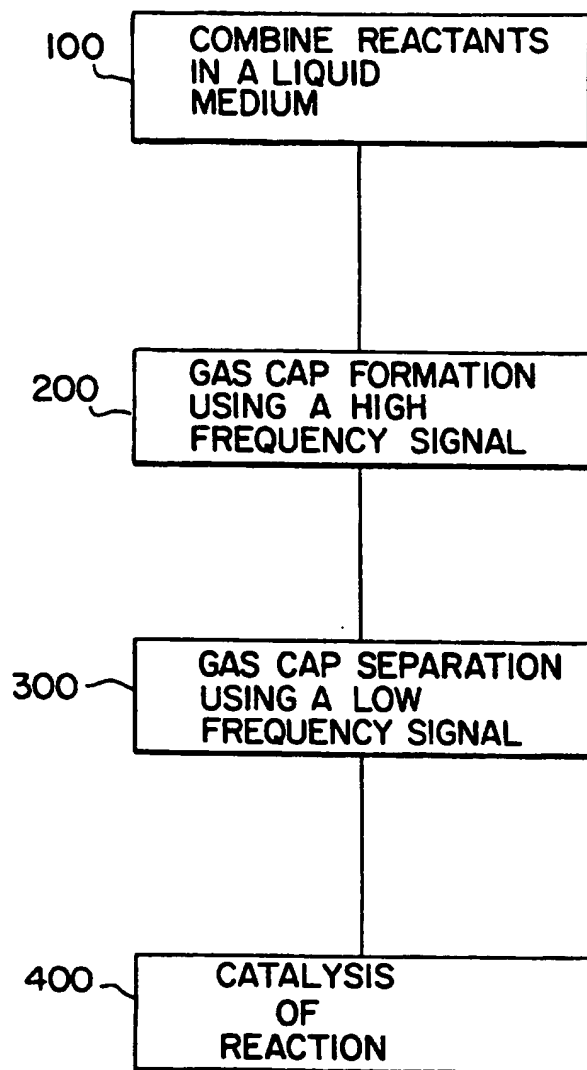


FIG. 1

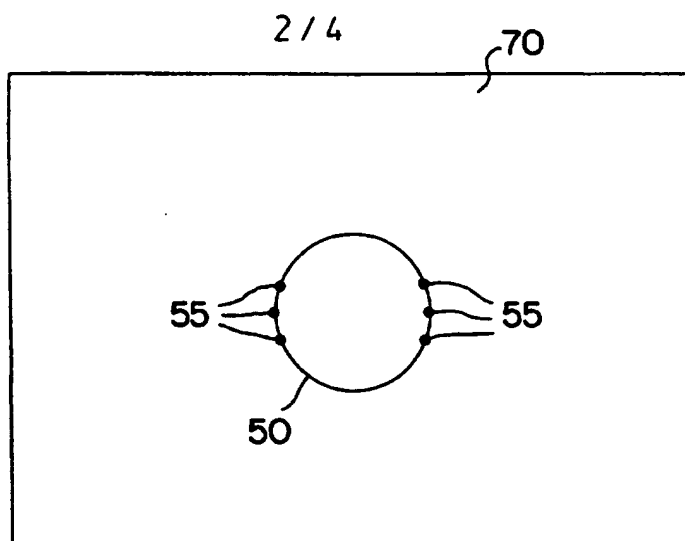


FIG. 2a

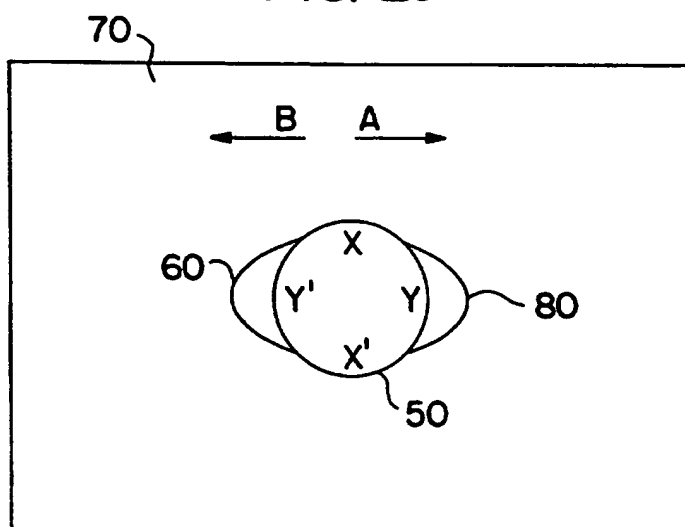


FIG. 2b

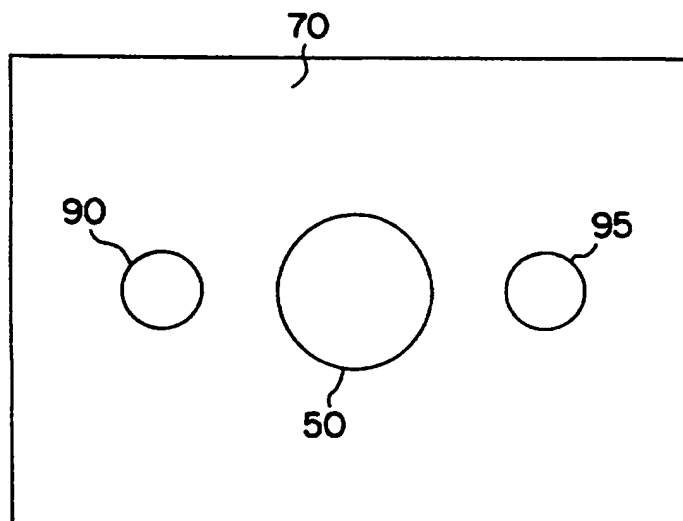


FIG. 2c

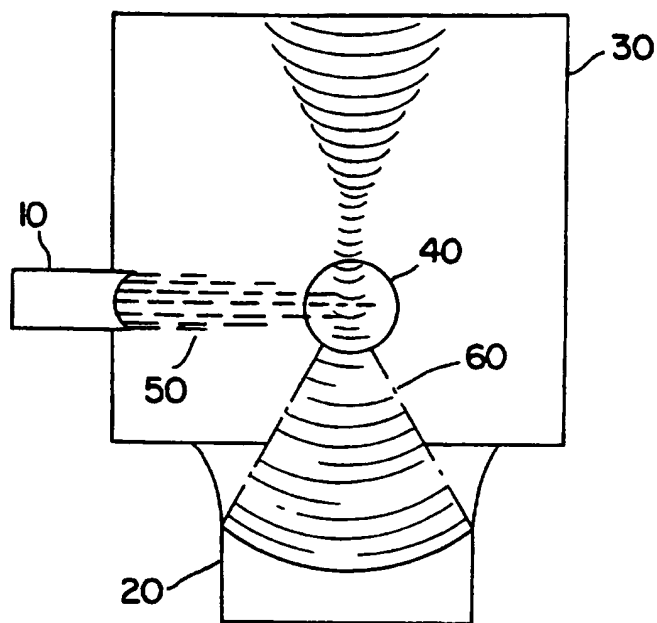


FIG. 3

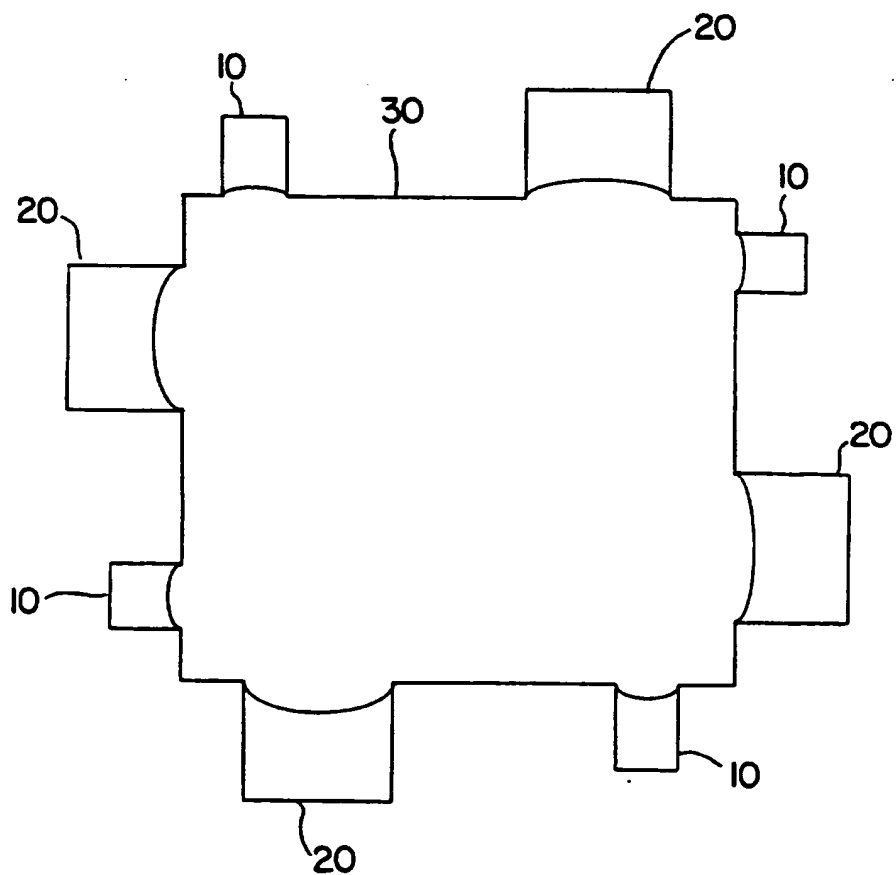


FIG. 4

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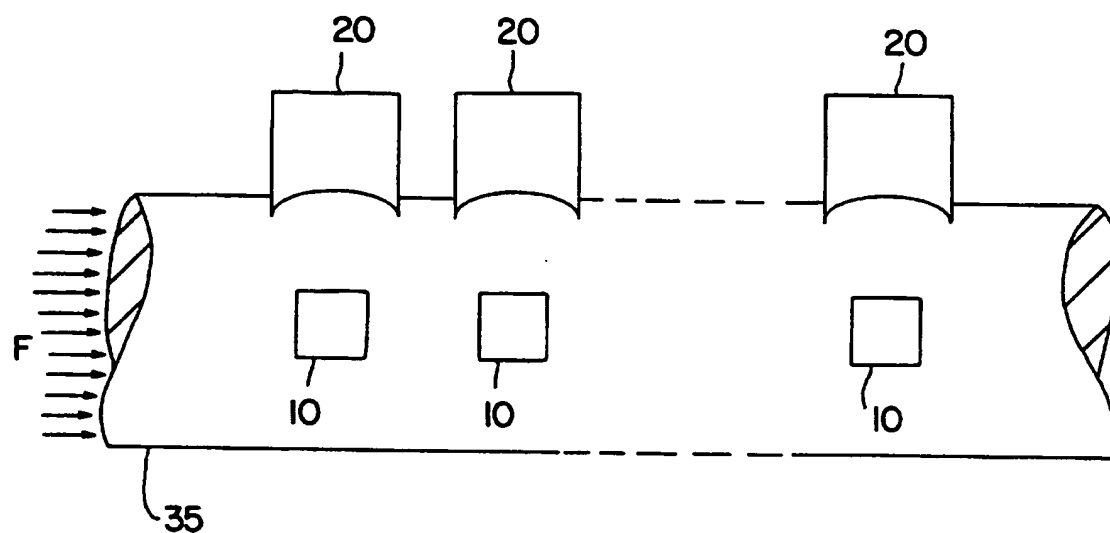


FIG. 5a

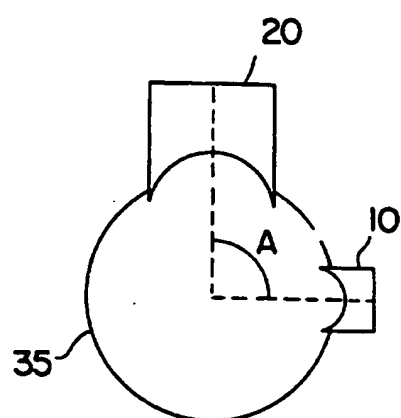


FIG. 5b

INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 96/00466

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 B01J19/10

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 B01J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US,A,4 369 100 (H.T.SAWYER) 18 January 1983 see abstract see column 2, line 41 - column 3, line 2 see column 3, line 38 - column 4, line 51 see column 5, line 65 - column 7, line 30 see column 8, line 3 - column 9, line 2 see column 9, line 58 - column 11, line 7 see column 12, line 4 - line 18 see figures	1,2,10, 11,19
A	---	3-9, 12-18
A	US,A,3 614 069 (FIBRA-SONICS, INC.) 19 October 1971 see the whole document ---	1-19
	-/--	

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Date of the actual completion of the international search

15 May 1996

Date of mailing of the international search report

28-05-1996

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INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	BE,A,1 005 386 (UNDATIM ULTRASONICS S.A.) 13 July 1993 see the whole document ---	1-19
A	FR,A,819 095 (I.G. FARBENINDUSTRIE AKTIENGESELLSCHAFT) 9 October 1937 see the whole document ---	1,10
A	CHEMICAL ENGINEERING, vol. 99, no. 10, October 1992, NEW YORK US, pages 37-41, XP002003113 W.GRINTHAL & G. ONDREY: "ULTRASOUND - SEEN BUT NOT HEARD" see the whole document ---	1-19
A	US,A,2 578 505 (SPERRY PRODUCTS, INC.) 11 December 1951 see the whole document ---	10,14,19
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T	RUSSIAN JOURNAL OF PHYSICAL CHEMISTRY, vol. 59, no. 10, 1985, NOTTINGHAM GB, pages 1569-1571, XP002003114 A.F.DMITRIEVA & M.A.MARGULIS: "The Simultaneous Effect of Two Acoustic Vibration Frequencies on the Rates of Sonochemical Reactions" see the whole document -----	

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 96/00466

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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BE-A-1005386	13-07-93	NONE	
FR-A-819095	11-10-37	NONE	
US-A-2578505	11-12-51	NONE	
US-A-2585103	12-02-52	NONE	

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